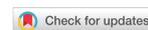


Research Article

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# Concentrations and human exposure to hexabromocyclododecane and tetrabromobisphenol A from the indoor environment in Bangkok metropolitan area, Thailand

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## Abstract

**Aim:** This study investigated hexabromocyclododecane (HBCDD) and tetrabromobisphenol A (TBBPA) concentrations in indoor dust from houses, offices, and cars and estimated toddler and adult exposure to HBCDD and TBBPA through dust ingestion.

**Methods:** The concentrations of HBCDD and TBBPA were measured in 47 indoor dust samples collected from the Bangkok metropolitan area, Thailand. All samples were analyzed for HBCDD and TBBPA using LC-MS/MS. The estimated daily intake (EDI) through dust ingestion was calculated from the median and 95th percentile concentrations of HBCDD and TBBPA.



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**Results:** HBCDD was detected in 47% of samples, and TBBPA was detected in all samples. The median concentrations of HBCDD were  $6.7 \text{ ng g}^{-1}$ ,  $<0.7 \text{ ng g}^{-1}$ , and  $<0.7 \text{ ng g}^{-1}$  in cars, houses, and offices, respectively. The isomer composition of  $\Sigma\text{HBCDD}$  in dust was:  $\alpha\text{-HBCDD}$  (40%-54%),  $\gamma\text{-HBCDD}$  (19%-40%), and  $\beta\text{-HBCDD}$  (17%-28%). In contrast, TBBPA was observed at higher concentrations, with median values of 674, 67, and  $22 \text{ ng g}^{-1}$  in offices, houses, and cars, respectively. Under a median exposure scenario, toddlers were exposed to  $0.05 \text{ ng kg}^{-1}\text{bw day}^{-1}$  for HBCDD and  $0.25 \text{ ng kg}^{-1}\text{bw day}^{-1}$  for TBBPA, with adults exposed to  $0.01$  and  $0.06 \text{ ng kg}^{-1}\text{bw day}^{-1}$  for HBCDD and TBBPA respectively.

**Conclusion:** Concentrations of HBCDD in dust from Thai cars, homes, and offices are lower than those of TBBPA following the listing of HBCDD in the Stockholm Convention on Persistent Organic Pollutants and limited use of HBCDD in Thailand in applications such as building insulation foam. Concentrations of TBBPA in office dust significantly exceeded ( $P < 0.05$ ) those in house and car dust owing to the greater number of electronic appliances and poor natural ventilation in offices. EDIs for Thai toddlers exceeded those of adults under both median and high-end exposure scenarios. However, EDIs of HBCDD and TBBPA for the general Thai population were below the corresponding oral reference dose guidelines.

**Keywords:** Hexabromocyclododecane, tetrabromobisphenol A, indoor environment, houses, offices, cars, daily intake, human exposure

## INTRODUCTION

Indoor environments are prominent sources of chemicals that contribute significantly to total human exposure because people spend most of their day indoors. Indoor dust is of great interest because it is omnipresent and acts as a repository for chemicals<sup>[1]</sup>, such as hexabromocyclododecane (HBCDD) and tetrabromobisphenol A (TBBPA). Thus, humans can be exposed to indoor pollutants<sup>[2,3]</sup>, which may cause significant adverse impacts on human health<sup>[4]</sup>. HBCDD is a brominated flame retardant (BFR) that is primarily used in building insulation material made from expanded and extruded polystyrene foam, as well as a back-coating for synthetic fabrics used as covers for sofas, chairs, etc. Minor uses of HBCDD are in high-impact polystyrene (HIPS) used in electrical and electronic appliances<sup>[5-7]</sup>. In addition to displaying environmental persistence and bioaccumulation potential, HBCDD can disrupt the thyroid and reproductive systems and affect the nervous and reproductive systems<sup>[3,8-9]</sup> and has thus been listed under Annex A of the Stockholm Convention on persistent organic pollutants since 2013<sup>[10]</sup>.

TBBPA is predominantly used as a reactive flame retardant in resins for application on printed circuit boards. However, it is used - albeit to a relatively minor extent - as an additive flame retardant in acrylonitrile butadiene styrene (ABS) and high-impact polystyrene. As additive FRs are not covalently bound to polymer components, they can be emitted more readily than reactive FRs from products and contaminate various environmental media<sup>[11-14]</sup>. This is of potential concern, as adverse effects of exposure to TBBPA include the disruption of the endocrine and thyroid hormone systems and neurotoxicity<sup>[6,14]</sup>. HBCDD and TBBPA are released into the environment during initial manufacturing and migration from consumer products and building materials, as well as during their recycling and disposal<sup>[11,15,16]</sup>. HBCDD and TBBPA are of increasing concern because of their chemical properties, such as low water solubility and low vapor pressure. Thus, they are persistent, transported long distances in the environment, highly toxic, and bioaccumulative<sup>[11,17]</sup>.

Indoor dust is a significant source of human exposure to HBCDD and TBBPA, especially for toddlers and children, since dust associated with indoor pollutants can enter the body through ingestion from hand-to-mouth behavior<sup>[6-8,18-20]</sup>. Moreover, indoor pollution is important for human health since most people spend

most of their time indoors<sup>[2]</sup>. Thus, toddlers or young children are more likely to be exposed to these substances through dust ingestion than adults<sup>[3,6,8]</sup>. Previous studies have revealed the presence of both HBCDD and TBBPA in indoor dust from several countries. House dust in the UK was contaminated at a concentration of 570,000 ng g<sup>-1</sup> of HBCDD<sup>[21]</sup> and offices in France were contaminated at a concentration of 10,188 ng g<sup>-1</sup><sup>[5]</sup>. TBBPA was present at 7260 ng g<sup>-1</sup> in houses in the USA<sup>[22]</sup> and 7951 ng g<sup>-1</sup> in Korean offices<sup>[6]</sup>. Notwithstanding these findings, there are, to our knowledge, few data on this issue in Southeast Asia and no data on concentrations of HBCDD and TBBPA in house dust from Thailand. Therefore, this article reports concentrations of HBCDD and TBBPA in floor dust collected from houses, offices, and cars in Bangkok. These concentrations are compared with those reported previously for other countries and used to evaluate human exposure to HBCDD and TBBPA through dust ingestion for Thai toddlers and adults.

## EXPERIMENTAL

### Chemicals and reagents

Solvents used for sample extraction and clean-up processes and analysis (e.g., hexane, dichloromethane, and methanol) were all HPLC grade chemicals purchased from Merck (Darmstadt, Germany). Concentrated sulfuric acid (98% purity) was obtained from Merck (Darmstadt, Germany). Standard solutions of  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HBCDD and TBBPA and isotopically labeled HBCDD, including <sup>13</sup>C<sub>12</sub>- $\alpha$ -HBCDD, <sup>13</sup>C<sub>12</sub>- $\beta$ -HBCDD, <sup>13</sup>C<sub>12</sub>- $\gamma$ -HBCDD, and <sup>13</sup>C<sub>12</sub>-TBBPA (each with a purity  $\geq$  98%), were purchased from Cambridge Isotope Laboratories (Andover, MA, USA). Standard d<sub>18</sub>- $\gamma$ -HBCDD was acquired from Wellington Laboratories (Guelph, ON, Canada). Silica gel 60 (0.063-0.200 mm) was obtained from Merck (Darmstadt, Germany). The indoor dust reference material SRM 2585 was supplied by the U.S. National Institute of Standards and Technology (NIST, Gaithersburg, MD, USA).

### Sample collection

Dust samples ( $n = 47$ ) were collected between November 2020 and January 2021 from the Bangkok metropolitan area in Thailand. Samples were collected from three categories of indoor microenvironments: houses ( $n = 20$ ), offices ( $n = 5$ ), and cars ( $n = 22$ ). The house and office dust sampling procedure was conducted according to previously reported protocols<sup>[5,17,21,23]</sup>. Briefly, in living rooms and offices, for floor dust, 1 m<sup>2</sup> of carpeted flooring was vacuumed for 2 min, while, for bare floors, 4 m<sup>2</sup> was sampled for 4 min. The sampling procedure in cars was based on a previous study<sup>[5]</sup>. Car dust samples were collected from private cars with an engine of 1550 cc -3000 cc. Most of the cars were Asian brands, and the age of the car ranged from 1 month to 20 years (average age of the car, 8 years  $\pm$  6 years). Dust samples were vacuumed from the surface of the seat and dashboard for 2 min. All samples were collected using a nylon sock with a 25  $\mu$ m pore size inserted into the nozzle of a portable vacuum cleaner tube. After each sampling, the sock was closed, wrapped with aluminum foil, and sealed in a plastic Ziplock bag. Samples were then placed in a clean glass container box and transported in a cooler with ice to the laboratory. The vacuum cleaner tube was cleaned thoroughly with water and an isopropanol-impregnated disposable wipe to prevent contamination. In the laboratory, each collected dust sample was passed through a pre-cleaned 250  $\mu$ m mesh to remove coarse particles, packed in clean aluminum foil, sealed in plastic Ziplock bags, and stored at -20 °C until analysis.

### Sample extraction and clean-up

Dust samples were extracted using a previously described method<sup>[17,21,23]</sup>. Before extraction, approximately 0.1 g of the sieved dust sample was spiked with 25 ng of each internal (or surrogate) standard (<sup>13</sup>C<sub>12</sub>- $\alpha$ -HBCDD, <sup>13</sup>C<sub>12</sub>- $\beta$ -HBCDD, <sup>13</sup>C<sub>12</sub>- $\gamma$ -HBCDD, and <sup>13</sup>C<sub>12</sub>-TBBPA). Then, 7 mL of hexane:dichloromethane mixture (1:1, v/v) was added to each sample. The sample was vortexed for 5 min and sonicated in an ultrasonic bath at 20 °C for 30 min. The resultant extracts were centrifuged for 5 min at 3500 rpm to

separate the supernatant and residual sample. The extraction process was repeated thrice. The combined supernatants were reduced to approximately 0.5 mL under a gentle nitrogen gas flow and reconstituted with 1 mL of hexane. The sample extract was then treated with concentrated sulfuric acid. The extracts were purified for the clean-up process using an SPE cartridge packed with 4 g of pre-cleaned acidified silica (44% concentrated sulfuric acid, w/w). Then, 3 mL of hexane:dichloromethane mixture (1:1, v/v) was used to pre-condition the cartridge and then discarded. After that, the extract (1 mL) was loaded onto cartridges with 2 mL × 1 mL hexane rinses. Cartridges were eluted with 25 mL of hexane:dichloromethane mixture (1:1, v/v). After the clean-up process, eluates were evaporated using a stream of nitrogen and reconstituted in 200 µL of methanol containing 25 pg/µL  $d_{18}$ - $\gamma$ -HBCDD as a recovery determination (or syringe) standard for LC-MS/MS analysis.

### Instrumental analysis

Instrumental analysis was performed using an Agilent 1200SL HPLC system coupled with an Agilent 6400 tandem mass spectrometer. HBCDD isomers ( $\alpha$ -,  $\beta$ -, and  $\gamma$ -HBCDD) and TBBPA were separated on an Agilent Pursuit XRS3 C18 reversed-phase analytical column (150 mm × 2.0 mm i.d., 3 µm particle size) maintained at 40 °C. The mobile phase of (A) 1:1 methanol/water and (B) methanol at a flow rate of 0.15 mL/min were applied to elute the target compound and 10 µL of each sample was injected. Mass spectrometry was performed in electrospray ionization negative mode. Nebulizer pressure was set at 50 psi and capillary voltage at 3500 V. The drying gas (nitrogen) was used at a flow rate of 10 L/min and set to 300 °C. MRM mode was used based on  $m/z$  640.4-78.8 and  $m/z$  652.4-79 for native and  $^{13}\text{C}$ -HBCDD labeled diastereomers, respectively. HBCDD isomers were baseline separated with retention times of 14.0, 14.6, and 15.0 min for  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HBCDD, respectively. For TBBPA, the quantitative determination of MRM was  $m/z$  540.8-78.8 for native and  $m/z$  552.8-78.8 for  $^{13}\text{C}$ -TBBPA. The retention time of TBBPA was 11.1 min.

### Quality assurance/quality control

All glassware was cleaned, rinsed with distilled water, rinsed with solvent, and then oven-dried for 5 h prior to use. For every batch of ten samples, one laboratory procedural blank was used to check for contamination during extraction and purification. In addition, field blanks ( $n = 5$ ) were also analyzed. These were obtained by spreading anhydrous sodium sulfate on pre-cleaned clean floors, vacuuming, and passing through all analytical processes as per real samples. The target substances were not detected in any procedural or field blanks. For further quality assurance, the certified reference material SRM 2585 (organics in indoor dust;  $n = 5$ ) was analyzed to evaluate the accuracy and precision of our analytical procedure. The measured value ranged 80%-123% of the certified values with relative standard deviation values below 15%. The limits of quantification (LOQs) of each compound were determined using a signal-to-noise ratio (S/N) of 10:1. LOQ values were 0.7 ng  $\text{g}^{-1}$  and 0.1 ng  $\text{g}^{-1}$  for HBCDD and TBBPA, respectively. The recovery of internal standards added to dust samples ranged 76%-120%, with a mean value of 98% ± 13% for HBCDD. The TBBPA recovery value ranged 70%-122%, with a mean value of 89% ± 16%.

### Estimation of daily exposure

Estimated daily intake (EDI, ng  $\text{kg}^{-1}\text{bw day}^{-1}$ ) values for HBCDD and TBBPA through dust ingestion for toddlers and adults were evaluated under the median and high-end exposure scenarios. EDI values were calculated using Equations (1) and (2)<sup>[11,15,19]</sup>:

$$EDI_{\text{ingestion for toddlers}} = \frac{[(C_H F_H) + (C_C F_C)] \times IR}{BW} \quad (1)$$

$$EDI_{\text{ingestion for adults}} = \frac{[(C_H F_H) + (C_O F_O) + (C_C F_C)] \times IR}{BW} \quad (2)$$

where  $C_H$ ,  $C_O$ , and  $C_C$  are the concentrations of HBCDD and TBBPA ( $\text{ng g}^{-1}$ ) in houses, offices, and cars, respectively.  $F_H$ ,  $F_O$ , and  $F_C$  are the exposure fractions (fraction of time spent in houses, offices, and cars), and IR is the dust ingestion rate ( $\text{g day}^{-1}$ ). BW is the body weight (kg) assumed for toddlers (12 kg)<sup>[24-26]</sup> and adults (63 kg)<sup>[8,11,18]</sup>. We assumed 100% absorption of HBCDD and TBBPA. The median and high dust ingestion rates assumed were  $0.05 \text{ g day}^{-1}$  and  $0.2 \text{ g day}^{-1}$  for toddlers and  $0.02 \text{ g day}^{-1}$  and  $0.05 \text{ g day}^{-1}$  for adults, respectively<sup>[5,16,19,27]</sup>. The exposure fraction assumed for toddlers was 86.1% of their time spent at home and 4.1% in cars. The figures for adults were 63.8% at home, 22.3% in offices, and 4.1% in cars<sup>[5,19,20]</sup>.

### Statistical analyses

Descriptive statistics (minimum, maximum, mean, median, and standard deviation) were calculated using Microsoft Office Excel 2016. Statistical comparisons were performed using IBM SPSS version 21. The distribution of concentrations of both TBBPA and HBCDD in the dust samples was evaluated for normal distribution using the Shapiro-Wilk test. This showed that sample concentrations did not display normal distribution. Therefore, concentrations were log-transformed. After log-transformation, TBBPA data were normally distributed, and the differences in TBBPA concentrations among our three indoor environment categories were evaluated using one-way ANOVA of log-transformed data. However, even after log-transformation, concentrations of HBCDD in house, office, and car dust remained not normally distributed. Therefore, the non-parametric Kruskal-Wallis test was used to evaluate any significant differences in concentrations of HBCDD among car, house, and office dust. Statistical significance was set at  $P < 0.05$ . When conducting statistical analysis, we assumed a zero concentration where a given contaminant in a sample was below the LOQ.

## RESULTS

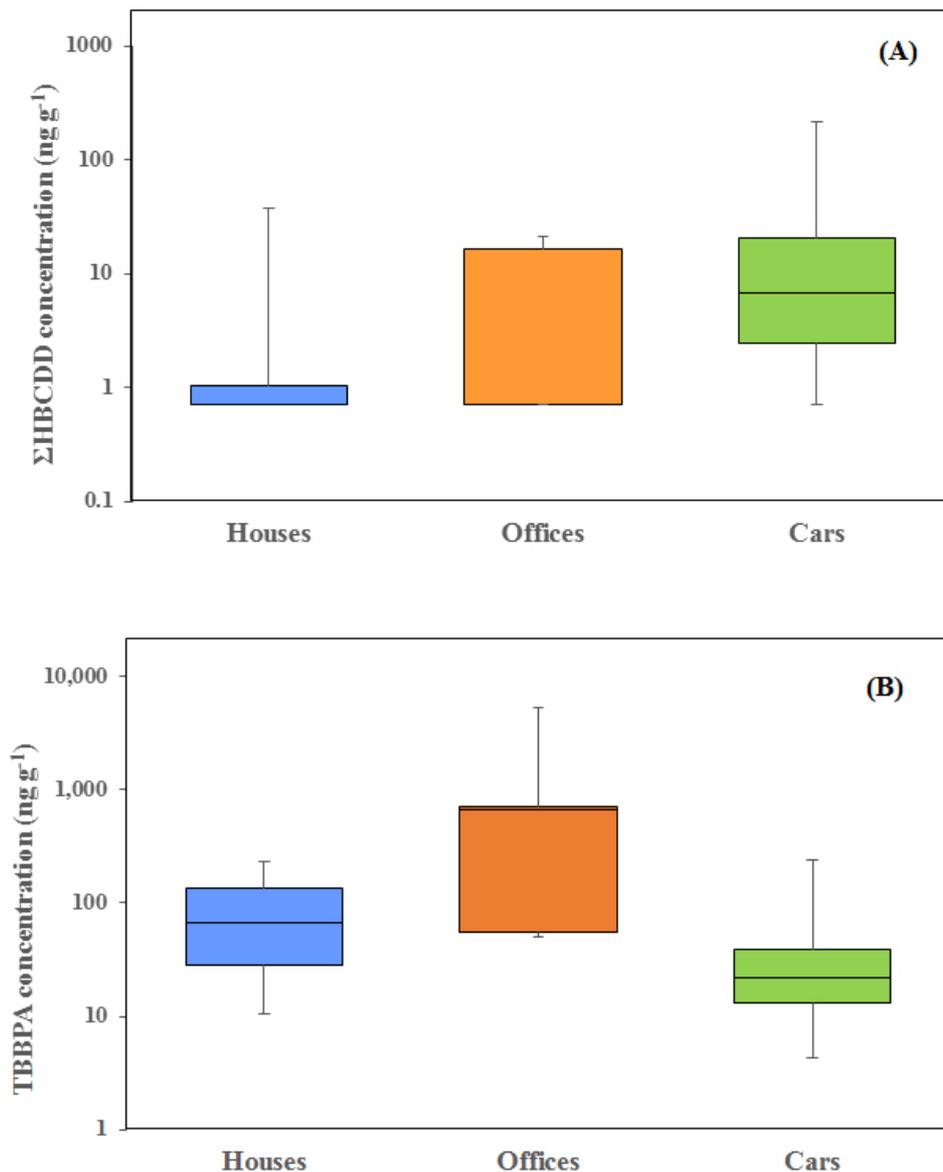
### Concentrations of HBCDD and TBBPA

HBCDD and TBBPA were measured in floor dust samples collected from three categories of indoor environment: houses, offices, and cars. HBCDD was detected in 22 of 47 samples, with detection frequencies of 17%, 40%, and 77% in houses, offices, and cars, respectively. Concentrations of  $\Sigma$ HBCDD (the sum of  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HBCDD isomers) in dust samples ranged from  $< 0.7 \text{ ng g}^{-1}$  to  $215 \text{ ng g}^{-1}$  (median  $6.7 \text{ ng g}^{-1}$ ) in cars,  $< 0.7 \text{ ng g}^{-1}$  to  $38 \text{ ng g}^{-1}$  (median  $< 0.7 \text{ ng g}^{-1}$ ) in houses, and  $< 0.7 \text{ ng g}^{-1}$  to  $21 \text{ ng g}^{-1}$  (median  $< 0.7 \text{ ng g}^{-1}$ ) in offices, as shown in Figure 1A. Concentrations were not significantly different among the three microenvironment categories ( $P = 0.658$ ). For the diastereomers of HBCDD, our study showed that the average isomer composition of  $\Sigma$ HBCDD in house dust samples was  $\alpha$ -HBCDD (42%),  $\beta$ -HBCDD (18%), and  $\gamma$ -HBCDD (40%). For office dust, the distribution was 52%, 20%, and 28% for  $\alpha$ -,  $\beta$ -, and  $\gamma$ -HBCDD, respectively. In car dust, the distribution was  $\alpha$ -HBCDD (54%),  $\beta$ -HBCDD (27%), and  $\gamma$ -HBCDD (19%) [Figure 2].

TBBPA was detected at concentrations above the LOQ in all samples. Median concentrations were  $22 \text{ ng g}^{-1}$  (4-242  $\text{ng g}^{-1}$ ),  $67 \text{ ng g}^{-1}$  (11-230  $\text{ng g}^{-1}$ ), and  $674 \text{ ng g}^{-1}$  (50-5319  $\text{ng g}^{-1}$ ) in car, house, and office dust, respectively [Figure 1B], with the concentration in office dust significantly exceeding those detected in house and car dust ( $P = 0.000$ ).

### Estimated daily intake of HBCDD and TBBPA

EDI values of HBCDD and TBBPA through dust ingestion for toddlers and adults living in the Bangkok metropolitan area, Thailand, were calculated based on the concentrations of these substances in house, office, and car dust. EDI values were calculated using both median and high-end exposure scenarios, as explained in the experimental section, based on ingestion of house and car dust for toddlers and house, car, and office dust for adults. Consistent with other studies elsewhere, these calculations revealed that toddlers had much higher EDIs of HBCDD and TBBPA than adults. The estimated median exposure to HBCDD was



**Figure 1.** Box plot of (A) HBCDD and (B) TBBPA concentrations in different indoor environments. The lower and upper boundaries of the boxes are the 25th and 75th percentiles. Error bars represent the minimum and maximum values. HBCDD: Hexabromocyclododecane; TBBPA: tetrabromobisphenol A.

0.05 ng kg<sup>-1</sup>·bw·day<sup>-1</sup> for toddlers and 0.01 ng·kg<sup>-1</sup>·bw·day<sup>-1</sup> for adults [Figure 3A], while, under the high-end exposure scenario, toddlers were exposed to 16 times more HBCDD. For TBBPA, the median EDI for toddlers was four times higher than for adults. High-end EDIs were 1.14 ng kg<sup>-1</sup> bw day<sup>-1</sup> and 0.89 ng kg<sup>-1</sup> bw day<sup>-1</sup> for toddlers and adults, respectively [Figure 3B].

## DISCUSSION

### Concentrations of HBCDD and TBBPA in indoor dust

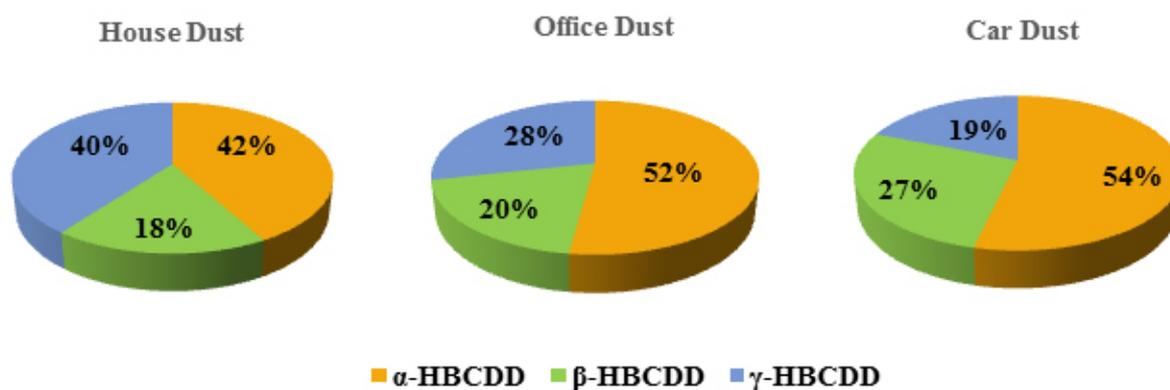
The concentrations of HBCDD and TBBPA detected in dust collected from houses, offices, and cars in studies reported previously for several countries are shown in Table 1. Generally, the median concentration of HBCDD in dust was highest in cars, followed by offices and houses (e.g., in France)<sup>[5]</sup>. However, in some

**Table 1. Median concentrations (ng g<sup>-1</sup>) of HBCDD and TBBPA from houses, offices, and cars in different countries**

Sample type	Location	Sampling year	Sample size (n)	α-HBCDD	β-HBCDD	γ-HBCDD	ΣHBCDD (Range)	TBBPA (Range)	Ref.
House dust	Thailand	2019 and 2020	20	< LOQ	< LOQ	< LOQ	< LOQ (< LOQ-38)	67 (11-230)	This study
	Spain	2016 and 2017	10	74.6	19.6	34.9	129 (12-1321)	-	Corsolini et al. <sup>[16]</sup> , 2021
	UK	2019	14	130	66	72	280 (76-570,000)	35 (< 0.5-71)	Drage et al. <sup>[21]</sup> , 2020
	Korea	2011	42	144	12	60	278 (< LOD-3,132)	69 (< LOD-2,092)	Kweon et al. <sup>[29]</sup> , 2018
	China	2014	20	-	-	-	34 (11-165)	5 (3-42)	Sun et al. <sup>[38]</sup> , 2018
	China	2014	30	64	21	64	156 (74-995)	20 (7-113)	Wang et al. <sup>[7]</sup> , 2018
	USA	2013	10	52	39	74	326 (104-636)	187 (0-7,260)	Allgood et al. <sup>[22]</sup> , 2017
	China	Not recorded (NR)	15	-	-	-	0.20 (0.08-1.4)	-	Peng et al. <sup>[19]</sup> , 2017
	Korea	2009 and 2016	46	43	6	46	106 (19-2645)	79 (14-1212)	Barghi et al. <sup>[6]</sup> , 2017
	Turkey	2012	10	-	-	-	251 (50-8800)	-	Kurt-Karakus et al. <sup>[1]</sup> , 2017
	South Africa	2012	7	-	-	-	-	120 (< 0.35-3767)	Abafe and Martincigh <sup>[11]</sup> , 2016
	France	2014	9	559	144	422	1125 (363-1865)	44 (7-165)	Abdallah et al. <sup>[5]</sup> , 2016
	Kazakhstan	2014	10	78	20	189	287 (112-450)	13 (< 0.06-83)	Abdallah et al. <sup>[5]</sup> , 2016
	Nigeria	2014	10	199	81	125	405 (41-1863)	50 (19-127)	Abdallah et al. <sup>[5]</sup> , 2016
	Egypt	2013	17	-	-	-	-	6.2 (1.4-153)	Hassan and Shoeib <sup>[15]</sup> , 2015
	USA	2012	30	7.9	27.8	70	338 (78-2528)	7.9 (< 0.2-245)	Stapleton et al. <sup>[28]</sup> , 2014
	Germany	2013	20	180	35	114	345 (53-4041)	28 (2.9-233)	Fromme et al. <sup>[8]</sup> , 2014
	USA	2011	16	62	16	73	160 (39-1800)	200 (22-2000)	Dodson et al. <sup>[32]</sup> , 2012
	New Zealand	NR	34	99	12	96	190 (20-4100)	-	Ali et al. <sup>[35]</sup> , 2012
	UK	2006 and 2007	45	380	93	670	1300 (140-140,000)	62 (< MQL-382)	Abdallah et al. <sup>[17]</sup> , 2008
Office	Thailand	2019 and 2020	5	< LOQ	< LOQ	< LOQ	< LOQ (< LOQ-21)	674 (50-5319)	This study
	China	2014	20	-	-	-	59 (18-224)	9 (2-14)	Sun et al. <sup>[38]</sup> , 2018
	China	2014	27	103	35	258	40 (8-171)	40 (8-171)	Wang et al. <sup>[7]</sup> , 2018
	Korea	2009 and 2016	18	137	34	294	496 (117-2519)	464 (138-7951)	Barghi et al. <sup>[6]</sup> , 2017
	Turkey	2012	9	-	-	-	424 (0.25-94,000)	-	Kurt-Karakus et al. <sup>[1]</sup> , 2017
	South Africa	2012	7	-	-	-	-	492 (< 1.15-2063)	Abafe and Martincigh <sup>[11]</sup> , 2016
	France	2014	11	2722	442	1329	4493 (1069-10188)	79 (32-1255)	Abdallah et al. <sup>[5]</sup> , 2016
	Kazakhstan	2014	10	106	32	123	261 (195-440)	4 (< 0.06-30)	Abdallah et al. <sup>[5]</sup> , 2016
	Nigeria	2014	10	76	28	55	159 (62-943)	30 (< 0.06-149)	Abdallah et al. <sup>[5]</sup> , 2016
	UK	2006 and 2007	28	220	84	470	760 (90-6600)	36 (< 0.05-140)	Abdallah et al. <sup>[17]</sup> , 2008

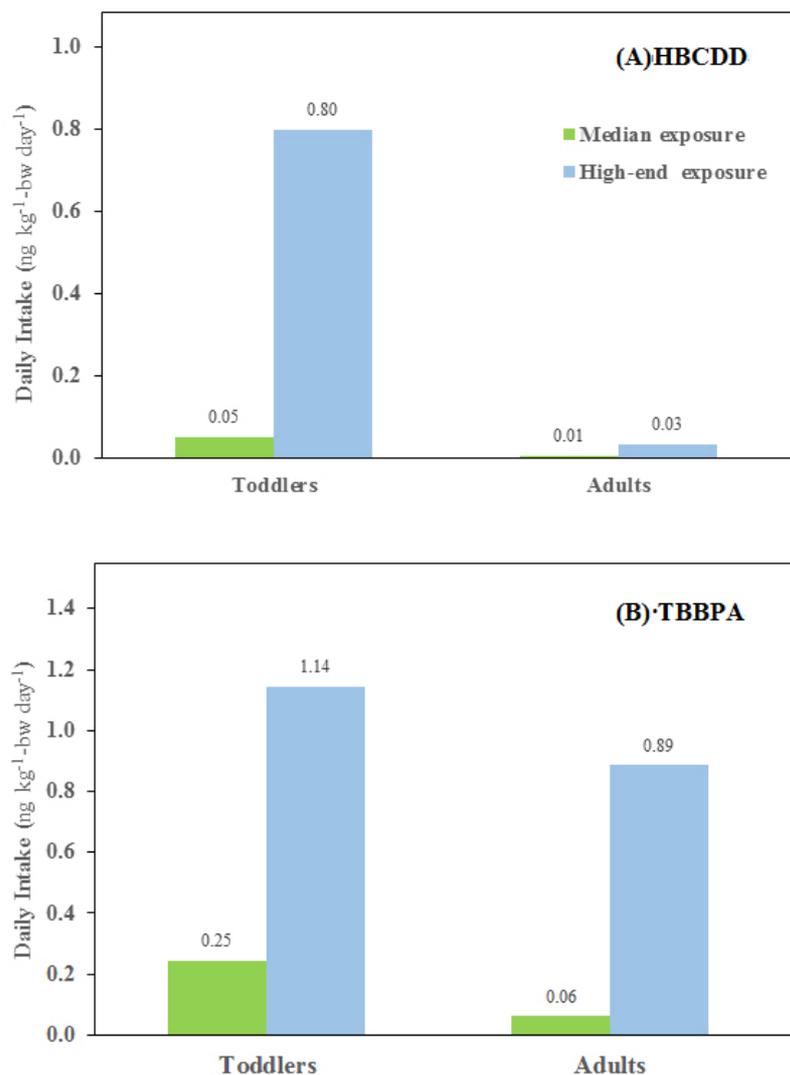
Cars	Thailand	2019 and 2020	22	2.68	0.88	2.09	6.7 (< LOQ-215)	22 (4-242)	This study
	Korea	2009 and 2016	19	185	21.41	98.98	297 (58-4172)	81 (46-651)	Barghi et al. <sup>[6]</sup> , 2017
	Greece	NR	30	90.3	15.8	46.4	155 (< LOQ-1745)	< LOQ (< LOQ-1064)	Besis et al. <sup>[18]</sup> , 2017
	China	NR	15	-	-	-	0.1 (0.05-0.3)	-	Peng et al. <sup>[19]</sup> , 2017
	France	2014	7	2221	629	1689	4539 (1458-7900)	47 (9-66)	Abdallah et al., 2016 <sup>[5]</sup>
	Kazakhstan	2014	11	609	231	1225	2065 (559-3962)	< 0.06 (< 0.06-5)	Abdallah et al. <sup>[5]</sup> , 2016
	Nigeria	2014	10	123	65	107	295	< 0.06 (< 0.06-6)	Abdallah et al. <sup>[5]</sup> , 2016
	South Africa	2012	14	-	-	-	-	1156 (< LOQ-4578)	Abafe and Martincigh <sup>[11]</sup> , 2016
	Egypt	2013	9	-	-	-	38 (17.39-105)	-	Hassan and Shoeib <sup>[15]</sup> , 2015
	Czech Republic	2008	25	< 0.3-275	< 0.3-57.3	< 0.3-739.5	92.6 (< 0.3-949.5)	-	Kalachova et al. <sup>[31]</sup> , 2012
	UK	2006 and 2007	20	2000	740	9600	13,000 (9190-69,000)	8 (< LOQ-25)	Abdallah et al. <sup>[17]</sup> , 2008

HBCDD: Hexabromocyclododecane; TBBPA: tetrabromobisphenol A; LOQ: limits of quantification.



**Figure 2.** Relative percent contribution of individual HBCDD isomers to the average concentration of  $\Sigma$ HBCDD in indoor dust from houses, offices, and cars in the Bangkok metropolitan area, Thailand. HBCDD: Hexabromocyclododecane.

countries, HBCDD concentrations were highest in office dust<sup>[1,6]</sup>. Of note is that the median HBCDD concentration of car dust in our study was lower than that reported for other countries. Median concentrations of HBCDD in car dust from the UK<sup>[17]</sup>, France<sup>[5]</sup>, Kazakhstan<sup>[5]</sup>, and Korea<sup>[6]</sup> were 13,000, 4539, 2065, and 297 ng·g<sup>-1</sup>, respectively, compared to 6.7 ng·g<sup>-1</sup> in our study. Overall, concentrations of HBCDD in our study of car, house, and office dust in Thailand were among the lowest globally<sup>[5,6,16,21-22,28-29]</sup>. This is likely because HBCDD usage in Thailand is reported to be low<sup>[30]</sup>. In general, the main diastereomer in HBCDD commercial mixtures is  $\gamma$ -HBCDD with smaller amounts of  $\alpha$ -HBCDD and  $\beta$ -HBCDD<sup>[31]</sup>. However, our study showed that the composition of  $\Sigma$ HBCDD in Thai dust was  $\alpha$ -HBCDD (40%-54%), followed by  $\gamma$ -HBCDD (19%-40%) and  $\beta$ -HBCDD (17%-28%). This HBCDD isomer profile is similar to that previously reported in indoor dust from other countries. Previous studies have highlighted the photolytically induced conversion of  $\gamma$ -HBCDD to  $\alpha$ -HBCDD in dust samples<sup>[5-7,18,19,31]</sup>.



**Figure 3.** Estimated daily exposure dose to (A) HBCDD and (B) TBBPA through indoor dust under the median and high-end exposure scenarios. HBCDD: Hexabromocyclododecane; TBBPA: tetrabromobisphenol A.

For TBBPA, previous studies from Korea<sup>[6]</sup> and South Africa<sup>[11]</sup> reported a high median concentration in office dust, similar to our study [Table 1]. Considering the use of TBBPA as an additive in HIPS and ABS resin in the plastic casings of electrical and electronic items<sup>[11]</sup>, the significantly higher concentrations detected in office compared to house and car dust in our study is likely because offices contain a greater number of electrical and electronic devices (computers, printers, photocopiers, etc.)<sup>[6,7,11]</sup>. For house dust, three previous studies recorded median concentrations of TBBPA in Korean ( $79 \text{ ng}\cdot\text{g}^{-1}$  and  $69 \text{ ng}\cdot\text{g}^{-1}$ )<sup>[6,29]</sup> and UK ( $62 \text{ ng}\cdot\text{g}^{-1}$ )<sup>[17]</sup> houses that are similar to the median concentration of  $67 \text{ ng}\cdot\text{g}^{-1}$  in our study. The concentration of TBBPA in Thai houses exceeded that in dust samples collected in Germany<sup>[8]</sup>, China<sup>[7]</sup>, and in a recent study in the UK<sup>[21]</sup>. In contrast, the median concentration of TBBPA in Thai houses was two and three times lower than that in South African<sup>[11]</sup> and American<sup>[22,32]</sup> houses, respectively.

In our study, the TBBPA concentration in dust from cars ranged from  $4 \text{ ng}\cdot\text{g}^{-1}$  to  $242 \text{ ng}\cdot\text{g}^{-1}$  (median  $22 \text{ ng}\cdot\text{g}^{-1}$ ). This median concentration of TBBPA in Thai car dust exceeded those reported for Greece<sup>[18]</sup>,

Nigeria<sup>[5]</sup>, Kazakhstan<sup>[5]</sup>, and the UK<sup>[17]</sup>. However, three studies from France<sup>[5]</sup>, Korea<sup>[6]</sup>, and South Africa<sup>[11]</sup> showed a higher median concentration of TBBPA than our results for Thai car dust. For example, Abafe and Martincigh<sup>[11]</sup> reported a median level of TBBPA in South African car dust of 1156 ng g<sup>-1</sup>, which was > 50 times higher than that reported in this study. Because they collected samples from resale and private cars, they explained that the elevated concentrations observed were due to the addition of TBBPA to ABS resin, which is mostly used as an automotive trim component and in automobile radios in resale cars. Moreover, the sampling period was during summer when outdoor temperatures were high, thereby facilitating TBBPA volatilization from in-car sources. In contrast, most of the car samples in our study were privately owned vehicles from which dust samples were collected during the evening in the cold season. In addition, many cars sampled were parked in garages with tinted films to filter sunlight. This combination of factors may have minimized the extent of any volatile emissions of TBBPA in cars in our study.

### Estimated daily intake of HBCDD and TBBPA

Reassuringly, even under high-end exposure scenarios, the EDI of HBCDD for both adults and toddlers was substantially below the oral reference dose (RfD) guideline value predicted to be toxic to the liver (200,000 ng·kg<sup>-1</sup>·bw·day<sup>-1</sup>) suggested by the US National Research Council<sup>[33-34]</sup>. The median exposure value in our result was similar to the EDIs of HBCDD from Egypt<sup>[15]</sup>, which were 0.03 and < 0.01 ng kg<sup>-1</sup> bw day<sup>-1</sup> for toddlers and adults, respectively [Table 2]. Furthermore, our results show lower EDIs than those in several previous studies. Briefly, the concentrations of HBCDD in dust from other global studies were higher than that in Thailand. Therefore, toddlers living in France<sup>[5]</sup>, the UK<sup>[17]</sup>, Korea<sup>[6]</sup>, Germany<sup>[8]</sup>, and New Zealand<sup>[35]</sup> were exposed to concentrations of HBCDD that were 170, 145, 96, 35, and 16 times higher, respectively, than Thai toddlers.

In our study, estimated TBBPA exposure of toddlers exceeded that of adults. However, the value of the high-end exposure scenario to TBBPA evaluated for toddlers and adults was lower than an RfD value (600,000 ng·kg<sup>-1</sup>·bw·day<sup>-1</sup>), proposed in 2015 by US authors based on uterine hyperplasia in rats<sup>[36]</sup>. However, some recent studies found that a lower dose of TBBPA in the environment (30,000 ng kg<sup>-1</sup> body weight) can affect health<sup>[37]</sup>. In comparison, Thai toddlers were exposed to TBBPA at concentrations three and two times higher than Chinese<sup>[38]</sup> and German<sup>[8]</sup> toddlers, respectively. This may be because the EDIs in the Chinese and German studies were based on house dust only. In contrast, our EDI was calculated based on dust from three indoor environment categories: houses, offices, and cars. In contrast, several previous studies, such as those in France<sup>[5]</sup>, the UK<sup>[17]</sup>, South Africa<sup>[11]</sup>, and Korea<sup>[6]</sup>, reported EDI values that exceed those of Thai toddlers. TBBPA concentrations in South Africa and Korea exceeded those in Thailand, although the EDIs in Korean studies included ingestion of dust from several indoor microenvironment categories, such as schools, which were not considered in our study. In addition, the body weight values were slightly different from those used in our study, resulting in the EDI values of these two countries exceeding those of Thailand.

## CONCLUSIONS

In this study, concentrations of HBCDD and TBBPA were measured in dust samples from houses, offices, and cars in the Bangkok metropolitan area of Thailand and used to estimate human exposure. The highest median concentration of HBCDD was observed in car dust (6.7 ng g<sup>-1</sup>), followed by houses and offices. Owing to the low application of HBCDD in Thailand in applications such as building insulation foam, concentrations of HBCDD in this study were among the lowest reported globally. In the case of TBBPA, the median concentration was highest in office dust (674 ng·g<sup>-1</sup>), followed by house dust (67 ng·g<sup>-1</sup>) and car (22 ng·g<sup>-1</sup>) dust. This may reflect a high density of electronic and electrical items used in offices. However, owing to the relatively small number of samples collected in this study, additional samples should be studied

**Table 2. Estimated daily intake (EDI) of HBCDD and TBBPA through dust ingestion of indoor environments in the Bangkok metropolitan area, Thailand**

Country	Type of dust	EDI of HBCDD (ng kg <sup>-1</sup> bw day <sup>-1</sup> )				EDI of TBBPA (ng kg <sup>-1</sup> bw day <sup>-1</sup> )				Ref.
		Toddler		Adults		Toddler		Adults		
		Median exposure	High-end exposure	Median exposure	High-end exposure	Median exposure	High-end exposure	Median exposure	High-end exposure	
Thailand	House, office, and cars	0.05	0.80	0.01	0.03	0.25	1.14	0.06	0.89	This study
Spain	House and laboratories	-	-	0.11	0.25	-	-	-	-	Corsolini et al. <sup>[16]</sup> , 2021
	House, laboratories, and computer room	-	-	0.24	0.61	-	-	-	-	Corsolini et al. <sup>[16]</sup> , 2021
China	Houses	0.43	-	0.04	-	0.09	-	0.01	-	Sun et al. <sup>[38]</sup> , 2018
	office	-	-	0.03	-	-	-	0.004	-	Sun et al. <sup>[38]</sup> , 2018
Korea	House, office, school, and cars	4.79	7.91	0.90	1.01	0.61	1.32	0.10	0.13	Barghi et al. <sup>[6]</sup> , 2017
Turkey	House and office	0.95	3.46	0.02	43	-	-	-	-	Kurt-Karakus et al. <sup>[1]</sup> , 2017
South Africa	House, office, and cars	-	-	-	-	0.60	2.41	0.08	1.92	Abafe and Martincigh <sup>[11]</sup> , 2016
France	House, office, and cars	8.5	61.6	0.65	2.93	0.47	5.61	0.03	0.27	Abdallah et al. <sup>[5]</sup> , 2016
Kazakhstan	House, office, and cars	0.83	4.88	0.06	0.23	0.07	0.83	< 0.01	0.04	Abdallah et al. <sup>[5]</sup> , 2016
Nigeria	House, office, and cars	2.16	22.3	0.17	1.07	0.19	1.57	0.01	0.07	Abdallah et al. <sup>[5]</sup> , 2016
Egypt	House, office, and cars	0.03	1.2	0.003	0.05	-	-	-	-	Hassan and Shoeib <sup>[15]</sup> , 2015
Germany	House	1.73	8.91	0.01	0.05	0.14	0.53	0.01	0.05	Fromme et al. <sup>[8]</sup> , 2014
New Zealand	House	0.78	29.3	0.05	1.26	-	-	-	-	Ali et al. <sup>[35]</sup> , 2012
UK	House, office, and cars	7.24	20.9	0.52	1.29	0.28	1.13	0.02	0.05	Abdallah et al. <sup>[17]</sup> , 2008

HBCDD: Hexabromocyclododecane; TBBPA: tetrabromobisphenol A.

to determine exposure by the Thai population to these substances. Estimated daily intakes through dust ingestion under both median and high-end exposure scenarios showed that toddlers were more highly exposed than adults. Reassuringly, the daily intakes of HBCDD and TBBPA for the general Thai population were well below the oral RfDs suggested by the US National Research Council and Wikoff *et al.*<sup>[36]</sup> (2015), respectively. However, it is important to note that the RfD for HBCDD was calculated by the US National Academy of Sciences (NAS) using data from Zeller and Kirsch (1970), which is an unpublished subchronic study performed on rats in 1970. The NAS concluded that confidence in this RfD for HBCDD is low because of a lack of other subchronic and chronic studies. Moreover, recent research has revealed that

dysfunction of the liver is a health concern following exposure to TBBPA at 30,000 ng kg<sup>-1</sup> body weight<sup>[37]</sup>. Therefore, chronic effects induced by continuous exposure to TBBPA and its derivatives should be further investigated. Continued monitoring of human exposure to both HBCDD and TBBPA via various pathways is thus prudent, especially for developmentally sensitive age groups such as toddler

## DECLARATIONS

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### Authors' contributions

Made substantial contributions to the conceptualisation and design of the study, sample collection and analysis, performed data analysis and interpretation, provided content, and wrote and edited the manuscript for submission: Waiyarat S

Conceptualized and designed the study; data analysis; edited the manuscript; and provided administrative, technical, material support, and funding acquisition: Boontanon SK

Made a significant contribution to the methodology of sample analysis and validation of these procedures: Boontanon N

Made substantial contributions to the conceptualization, methodology of sample collection and analysis, and edited the manuscript: Harrad S, Abdallah MAE, Drage DS

### Availability of Data and Materials

Not applicable.

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### Conflicts of interest

All authors declared that there are no conflicts of interest.

### Ethical approval and consent to participate

Not applicable.

### Consent for publication

Not applicable.

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